

REMARKS

Applicants are presently amending their claims in order to further clarify the definition of various aspects of the present invention, and so as to facilitate proceedings in connection with the above-identified application. Specifically, Applicants are submitting new independent claim 14, directed to a process for producing coke. Claim 14 recites the subject matter set forth in each of claims 1, 5 and 7. In light of new claim 14, claims 1 and 5-7 are being cancelled without prejudice or disclaimer, and dependencies of claims 2 and 10-12 are being amended, to be dependent on claim 14.

In addition, the subject matter of claim 13 has been incorporated into claim 8; and, correspondingly, claim 13 has been cancelled without prejudice or disclaimer.

Initially, it is respectfully requested that the present amendments be entered, notwithstanding Finality of the Office Action mailed October 14, 2004. In this regard, noting that claim 14 is a combination of subject matter set forth in previously considered claims 1, 5 and 7, with claim 7 being dependent on previously considered claim 1, it is respectfully submitted that the present amendments to the process claims do not raise any new issues, including any issue of new matter. Moreover, further noting that the present amendments to the pitch composition claims set forth the subject matter of previously considered claim 13 in independent form, it is respectfully submitted that the present amendments to the pitch composition claims do not raise any new issues, including any issue of new matter. Noting that the present amendments further define and clarify the subject matter of various aspects of the present invention, including setting forth in independent claims the subject matter of previously considered claims, it is respectfully submitted that the present

amendments materially limit any issues remaining in connection with the above-identified application, and, at the very least, present the claims in better form for appeal. Furthermore, noting clarification and further explanation of the rejections as set forth in the Final Office Action mailed October 14, 2004, it is respectfully submitted that the present amendments are timely.

While the present amendments add new claim 14, it is to be noted that these amendments cancel claims 1, 5-7 and 13, and thus reduce the number of claims to be considered on the merits in the above-identified application, as compared to the number of finally rejected claims.

In view of all of the foregoing, it is respectfully submitted that Applicants have made the necessary showing under 37 CFR § 1.116(c); and that, accordingly, entry of the present amendments is clearly proper.

Applicants respectfully submit that all of the claims presented for consideration by the Examiner patentably distinguish over the teachings of the references applied by the Examiner in rejecting claims in the Office Action mailed October 14, 2004, that is, the teachings of the U.S. patents to Mochida, et al., No. 4,891,126, to Orac, et al., No. 5,843,298, to Hayner, No. 6,153,004, and to Ryu, et al., No. 6,521,380, and Japanese Patent Document No. 1-282346 to Matsumoto, under the provisions of 35 USC §103.

It is respectfully submitted that these references as applied by the Examiner would have neither taught nor would have suggested such a process for producing a carbon material for a negative electrode of a non-aqueous solvent type secondary battery as in the present claims, including, inter alia, mixing the specified amounts of the recited mesophase pitch (which is produced by polymerizing a condensed

polycyclic hydrocarbon or a substance containing the condensed polycyclic hydrocarbon in the presence of hydrogen fluoride-boron trifluoride), with coal tar pitch which contains substantially no quinoline insolubles, and further mixing specified amounts of sulfur, prior to heat-treating at a temperature of at least 500°C, pulverizing, and graphitizing at a temperature of at least 2000°C. See claim 14.

In addition, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such process as in the present claims, having features as discussed previously in connection with claim 14, and furthermore wherein the mesophase pitch has a softening point of 150°C or higher as measured by a specified test method and a carbonization yield of 70% or higher (see claim 2); and/or wherein 1-30 parts by weight sulfur per 100 parts by weight of the pitch composition obtained in step (1), is mixed with the pitch composition (see claim 10); and/or wherein the coke is pulverized to provide a coke powder, this coke powder being subjected to the graphitizing (see claim 11); and/or wherein the graphitizing forms a graphite powder (see claim 12).

Furthermore, it is respectfully submitted that the teachings of the applied prior art would have neither disclosed nor would have suggested a pitch composition as in the present claims, comprising specified amounts of mesophase pitch, coal tar pitch and sulfur, and having an optically anisotropic content of 1-99% by volume, and wherein the mesophase pitch is a pitch produced by polymerizing a condensed polycyclic hydrocarbon or a substance containing the condensed polycyclic hydrocarbon in the presence of hydrogen fluoride-boron trifluoride, and the coal tar pitch contains substantially no quinoline insoluble. See claim 8.

The present invention is directed to a process for producing a carbon material for a negative electrode of a non-aqueous solvent-type secondary battery, as well as compositions, e.g., useful in this process.

Mesophase pitch is an excellent carbon material capable of producing pitch coke having a high graphitizability at a high yield. However, when heat treating this mesophase pitch the pitch is foamed, such that the coke derived therefrom disadvantageously exhibits a low density. Such low density coke is disadvantageous when, e.g., used in forming negative electrodes of non-aqueous solvent secondary batteries. It has been desired to avoid such foaming, and various techniques therefor have been proposed, as discussed on pages 1 and 2 of Applicants' specification. However, these techniques have not been satisfactory in avoiding foaming, while also producing an artificial graphite having a high graphitization degree and high charge-discharge efficiency at initial cycles when this material is incorporated in a negative electrode of, e.g., a lithium ion secondary battery.

Against this background, Applicants provide a process for producing, from mesophase pitch, a carbon material for, e.g., a negative electrode of a non-aqueous solvent type lithium ion secondary battery, having a high discharge capacity and a high charge-discharge efficiency, which can be produced at a high productivity and while avoiding foaming. Applicants also provide a pitch composition which can be used in such process. Applicants have found that when a pitch mixture containing 100 parts by weight of mesophase pitch, has included therein 10-1000 parts by weight of coal tar pitch, this pitch mixture being heat-treated at a temperature of 500°C or higher, foaming of the pitch is avoided; and when the coke is graphitized at a temperature of 2000°C or higher, it is possible to obtain an artificial graphite having

a high graphitization degree. Especially advantageous results are achieved when specified mesophase pitch and coal tar pitch as in the present claims are used. Including sulfur in the pitch composition further inhibits foaming of the pitch and more effectively produces coke. Moreover, when the coke is pulverized, and graphitized at a temperature of 2000°C or higher, it is possible to obtain a high-crystallinity graphite powder which can be suitably used as a carbon material for a negative electrode of a non-aqueous solvent type secondary battery having a high discharge capacity and a high charge-discharge efficiency. Note the paragraph bridging pages 3 and 4 of Applicants' specification.

Thus, as one feature of the present invention, the pitch composition includes 10-1000 parts by weight of coal tar pitch, of the specified type, per 100 parts by weight of the mesophase pitch of the specified type. When the amount of the coal tar pitch mixed is less than 10 parts by weight, it is not possible to effectively prevent the pitch from being foamed. When the amount of the coal tar pitch mixed is more than 1000 parts by weight, the pitch is deteriorated in carbonization yield as well as graphitizability. Note the paragraph bridging pages 5 and 6 of Applicants' specification.

By mixing the coal tar pitch having a relatively low carbonization reaction rate with the mesophase pitch, viscosity of the reaction system is kept low until reaching a high temperature, thereby inhibiting the growth of foams. As a result, it becomes possible to produce the high density coke at a high yield without foaming. Note page 6, lines 16-25, of Applicants' specification.

As an additional feature according to the present invention, the pitch composition further includes sulfur in a specified amount, to further inhibit the pitch

from being foamed and more effectively produce coke. Note the paragraph bridging pages 6 and 7 of Applicants' specification.

Matsumoto discloses a technique for production of a pitch-based carbon fiber. The technique includes hot-melt spinning of a mixture of a coal-based mesophase pitch and a petroleum-based mesophase pitch, followed by insolubilization and firing. This patent document discloses that desirably the spinning mesophase pitch raw materials have a melting point of 220-300°C, toluene insoluble parts of at least 70 wt.%, and optical anisotropy of at least 80 vol%. See page 4 of the English translation of this Japanese patent document prepared at the U.S. Patent and Trademark Office. This patent document discloses that for blending the petroleum type and coal type mesophase pitches, pulverization mixing or hot-melt mixing can be used; and that for the carbonization treatment, a method where its heating is done to 800°-1700°C in an inert gas atmosphere or in a vacuum can be used, and for the graphitization treatment a method wherein heat treatment is applied at 1700°C or above in an inert atmosphere can be used. Note page 4 of this English translation.

It is respectfully submitted that the applied Japanese patent document is directed to production of pitch-based carbon fiber; and that in forming this fiber a blend of coal-based and petroleum-based mesophase pitches (that is, two different types of mesophase pitches) is used. It is respectfully submitted that this patent document would have neither taught nor would have suggested a pitch composition or process as in the present claims, including, inter alia, the coal tar pitch included with the mesophase pitch, and amounts thereof, and also wherein the mesophase pitch and coal tar pitch are those as recited in the present claims. In this regard, clearly the applied Japanese patent document would have neither disclosed nor

would have suggested such process or composition including, inter alia, the coal tar pitch containing substantially no quinoline insolubles, and/or the mesophase pitch produced by the specified process set forth in claims 1 and 8 as presently amended.

Furthermore, it is emphasized that the applied Japanese patent document discloses forming a carbon fiber, with hot-melt spinning being performed relatively early in the processing (that is, after forming the mixture), and thereafter carbonization and graphitization treatments being performed. It is respectfully submitted that the disclosure of this patent document would have taught away from processing as in the present invention, including, inter alia, pulverizing the coke and graphitizing the pulverized coke; and, in particular, wherein the coke is pulverized to provide a coke powder, the coke powder being subjected to the graphitizing, as in claim 11, or wherein the graphitizing forms a graphite powder, as in claim 12.

It is emphasized that the applied Japanese patent document discloses a blend of mesophase pitches, each pitch preferably having a melting point of 220°-300°C, more than 70 wt.% in toluene insolubles and more than 80 vol% optical anisotropy. Note that, e.g., illustratively disclosed therein is a composition of a coal-based mesophase pitch of 245°C melting point, 83 wt.% toluene insolubles and 85 vol% optical anisotropy, blended with a petroleum-based mesophase pitch of 250°C melting point, 79 wt.% toluene insolubles and 98 vol% optical anisotropy.

In contrast, according to the invention as presently claimed, a mesophase pitch is produced from a condensed polycyclic hydrocarbon in the presence of HF/BF₃ and is mixed with a coal tar pitch, which contains substantially no quinoline insolubles, and thereafter the mixture is subjected to heat-treatment and further treated to form pulverized coke, in producing a carbon material for a negative

electrode of a non-aqueous solvent type secondary battery. It is respectfully submitted that coal tar pitch used in connection with the present invention is ordinary coal tar pitch which contains substantially no quinoline insolubles. It is respectfully submitted that the coal tar pitch is isotropic and has an optical anisotropic content of 0%, as can be seen, for example, in Example 1 on pages 9 and 10 of Applicants' specification. Clearly, the blend of mesophase pitches as disclosed in the applied Japanese patent document would have neither taught nor would have suggested the presently claimed subject matter, including components of the pitch composition (including, e.g., the coal tar pitch), and advantages thereof as discussed in the foregoing.

It is emphasized that in Matsumoto, a coal-based mesophase pitch and a petroleum-based mesophase pitch are mixed to produce carbonized fiber and graphitized fiber while shortening insolubilization (oxidation reaction) time of the mesophase pitch fiber and preventing cracking in the cross-section of this fiber. Clearly, the objects and results of the present invention are quite different from that of Matsumoto.

Since a coal-based mesophase pitch and a petroleum-based mesophase pitch are mixed in Matsumoto, clearly the coal tar pitch used in the present invention is different from the coal-based mesophase pitch used in Matsumoto. Also, the petroleum-based mesophase pitch used in Matsumoto is different from the mesophase pitch produced by polymerizing a condensed polycyclic hydrocarbon or a substance containing the condensed polycyclic hydrocarbon in the presence of HF-BF₃, claimed in the present invention.

It is respectfully submitted that the secondary references applied by the Examiner would not have rectified the deficiencies of Matsumoto, such that the presently claimed invention as a whole would have been obvious to one of ordinary skill in the art.

Mochida, et al. discloses a mesophase pitch for use in the production of high-performance carbon fibers and other carbon materials. The pitch is produced by polymerizing a condensed polycyclic aromatic hydrocarbon or a substance that contains it, with the pitch having a hydrogen-to-carbon atomic ratio of from about 0.5 to about 1.0, containing naphthenic carbon in an amount of at least about 7% of the total carbon, and containing at least about 90% of an optically anisotropic phase. Note column 4, lines 40-48; see also column 4, lines 49-54, with respect to a process for producing this mesophase pitch. Note also column 5, lines 36-44 of this patent, describing starting materials for the production of the mesophase pitch; and column 6, lines 10-12, for the temperature for obtaining the mesophase pitch. See also column 7, lines 25-38, with respect to techniques for forming carbon fibers from the pitch.

It is respectfully submitted that Mochida, et al. discloses a specific pitch, useful illustratively, as set forth in the patent, for producing carbon fibers. It is respectfully submitted that Mochida, et al. and the applied Japanese patent document, either alone or in combination, would have neither taught nor would have suggested the materials mixed in the process for producing a carbon material for a negative electrode of a non-aqueous solvent type secondary battery, as in the present claims, or the materials of the pitch composition of the present claims, much less amounts of these materials as in the present claims.

Noting that Mochida, et al. is primarily concerned with forming fibers, it is respectfully submitted that the teachings of this reference together with the teachings of the applied Japanese patent document would have neither disclosed nor would have suggested, inter alia, the pulverizing and graphitizing the pulverized coke, as in the present process claims.

Orac, et al. is concerned with a method of distilling coal tar containing quinoline insolubles (QI) solids to provide coal-tar pitch having increased QI concentration and, concurrently, a QI-free coal-tar pitch. The process is described most generally at column 2, lines 35-56, and includes use of a cross-flow filtration membrane filter and a pump to circulate continuously heated dehydrated tar to obtain (i) a substantially QI-free permeate stream exiting a circulation loop via the cross-flow filter and (ii) a QI-containing stream. This patent further discloses that in production of graphite electrodes for the steel industry, a pitch impregnate is used to fill the pores generated during initial carbonization of the carbon article, to increase final graphite product strength and density, the impregnating pitches preferably being free of QI, the QI-free permeate stream formed by the process of this patent being usable as the pitch impregnate.

Initially, note that the applied Japanese patent document is concerned with production of carbon fiber, with Mochida, et al. being primarily concerned with fibers; in contrast, Orac, et al. is concerned with production of graphite electrodes for the steel industry. Different problems are addressed in the applied Japanese patent document (that is, production of fiber that is free from cracks on its cross section) and by Orac, et al. In view of the different technologies involved and different problems addressed, it is respectfully submitted that one of ordinary skill in the art

concerned with in the applied Japanese patent document would not have looked to the teachings of Orac, et al. In other words, it is respectfully submitted that these teachings are directed to non-analogous arts.

Furthermore, again noting the differences in technologies involved in the applied Japanese patent document and Mochida, et al., on the one hand, and Orac, et al., on the other, it is respectfully submitted that there would have been no motivation for combining the teachings of Orac, et al. with the teachings of the applied Japanese patent document and Mochida, et al.

The conclusion by the Examiner in the third paragraph on page 4 of the Office Action mailed October 14, 2004, that it would have been obvious to have modified the process of the applied Japanese patent document "by using the coal tar pitch from the process of Orac because, in the process [of the applied Japanese patent document], one of skill in the art would use any coal tar pitch including the pitch from the Orac process", is respectfully traversed. The Examiner has provided no basis for his conclusion that one of ordinary skill in the art would use "any coal tar pitch" in the process of the applied Japanese patent document, especially since the Japanese patent document discloses use of coal-based mesophase pitch; and in the absence of a basis for this conclusion, clearly the conclusion is improper.

The contention by the Examiner in the first full paragraph on page 8 of the Office Action mailed October 14, 2004, that one of skill in the art would use any coal tar pitch including the coal tar pitch of Orac, et al., in the material of Matsumoto, is respectfully traversed. In this regard, it is emphasized that the coal tar pitch in Orac, et al. is not a mesophase pitch, required in Matsumoto. It is respectfully submitted that the coal tar pitch described in Orac, et al. would not be used for the

production of carbonized fiber. The Examiner has pointed to no basis for his conclusion that one of ordinary skill in the art would use any coal tar pitch in the technique of Matsumoto. Clearly, without any evidence in support of this contention by the Examiner, such contention providing a basis for the conclusion of obviousness is improper. See In re McKellin, 188 USPQ 428 (CCPA 1976).

Furthermore, even assuming, arguendo, that the teachings of Orac, et al., Mochida, et al. and the applied Japanese patent document were properly combinable, such combined teachings would have neither disclosed nor would have suggested the presently claimed subject matter, including specific components of the pitch composition and amounts of these components, and optically anisotropic content, and advantages achieved by the present invention due thereto. In this regard, attention is again directed to the paragraph bridging pages 5 and 6 of Applicants' specification, showing unexpectedly better results achieved through using amounts of materials, and in particular amount of the coal tar pitch, as in the present claims. Particularly in view of these unexpectedly better results, the Examiner has not established obviousness of the presently claimed subject matter, including amounts of components of the pitch.

Hayner discloses asphalt compositions containing asphalt and sulfur, particularly for road building. This patent also discloses a method of preparing such compositions by forming a mixture of asphalt, sulfur and liquid hydrocarbon oil, which includes forming a sulfur slurry including solid sulfur and a liquid hydrocarbon oil and mixing this sulfur slurry with a polymer-free asphalt. Note column 4, lines 29-38.

Initially, it is noted that Hayner discloses forming asphalt compositions primarily for building of roads, which compositions include sulfur, the sulfur achieving

marked changes in the asphalt properties, e.g., used in road building. Specifically, Hayner discloses that a slurry of solid sulfur in oil, or oil containing sulfur compounds, is added to asphalt, to avoid uneven mixing which can occur when sulfur is separately added as a solid or in molten form, and also avoids formation of explosive clouds of sulfur dust, as described in the Abstract of Hayner.

Clearly, the technical field and problem addressed by the present invention is different from that of Hayner, and the effect of the present invention is quite different from that of Hayner. In the present invention, by adding sulfur to the mixture of special mesophase pitch and coal tar pitch in, e.g., the process for producing a carbon material for a negative electrode, foaming is further inhibited and a carbon material for a negative electrode of non-aqueous solvent type secondary batteries having a high discharge capacity and high charge-discharge efficiency is achieved, due to, e.g., the crystallite size being increased by adding the sulfur. Compare Examples 1 and 2 of Applicants' original disclosure on pages 9-12 of Applicants' specification, wherein when sulfur is added in Fig. 2 the crystallite size is greater than that in Example 1. It is respectfully submitted that the advantageous effects achieved according to the present invention would have neither been disclosed nor would have been suggested by the teachings of Hayner either alone or in combination with the teachings of the other applied references. Especially in view thereof, it is respectfully submitted that the presently claimed subject matter, including incorporation of sulfur, particularly in amounts as set forth in the present claims, would have neither been disclosed nor would have been suggested by the teachings of the applied references, including Hayner.

Furthermore, it is emphasized that Hayner is primarily concerned with forming asphalt compositions for building of roads. It is respectfully submitted that one of ordinary skill in the art concerned with in the applied Japanese patent document, directed to formation of fibers, would not have looked to the teachings of Hayner. That is, in view of the entirely different technologies involved, and different problems addressed by each, these documents are directed to non-analogous arts, such that one of ordinary skill in the art concerned with in the applied Japanese patent document would not have looked to the teachings of Hayner.

Moreover, it is respectfully submitted that there would have been no motivation for combining the teachings of Hayner with the teachings of, inter alia, the applied Japanese patent document.

The contention by the Examiner in the fourth full paragraph on page 5 of the Office Action mailed October 14, 2004, that it would have been obvious to add sulfur to the pitch composition of Matsumoto because adding sulfur to the pitch would increase viscosity and reduce the melting point of the pitch, is noted. The Examiner points to no evidence supporting his statement that it was known prior to the present invention that adding sulfur to the pitch "would increase viscosity and reduce the melting point of the pitch". In view of the lack of evidence, this conclusion by the Examiner as to motivation for combining the teachings of the references is clearly improper.

It is again emphasized that Hayner is concerned with adding sulfur to an asphalt blend, primarily for use in building roads. It is respectfully submitted that the teachings of Hayner as a whole must be considered; and, in light thereof, it is respectfully submitted that one of ordinary skill in the art concerned with in the

process of the applied Japanese patent document would not have looked to the teachings of Hayner. In this regard, the Examiner has not even alleged that the applied Japanese patent document would want “increased viscosity and reduced melting point” of the pitch, the motivation alleged by the Examiner for combining teachings of the references.

In any event, even assuming, arguendo, that the teachings of Hayner were properly combinable with the teachings of the other references as applied by the Examiner, such combined teachings would have neither disclosed nor would have suggested the presently claimed subject matter, including mixing the specified amount of sulfur as in the present claims, and advantages thereof.

Ryu, et al. discloses a rechargeable lithium battery, having a negative electrode including a graphite-based active material with boron as a donor and a positive electrode with a transition metal oxide-based active material, a separator being interposed between the negative and positive electrodes. The positive and negative electrodes, and separator, are all saturated with an electrolyte, this electrolyte containing at least 51% by volume of cyclic carbonate and chain carbonate of 49% by volume. See column 2, lines 8-16. See also column 3, lines 21-53, for various techniques for forming the negative electrode active material.

It is again emphasized that the applied Japanese patent document is directed to a carbon fiber. In view of the differences in technology in the applied Japanese patent document, on the one hand, and in Ryu, et al, on the other, it is respectfully submitted that one of ordinary skill in the art concerned with in the applied Japanese patent document would not have looked to the teachings of Ryu, et al. Furthermore, there would have been no motivation for combining the teachings of the applied

references, including the applied Japanese patent document, with the teachings of Ryu, et al.

Moreover, it is emphasized that the Examiner has applied Ryu, et al. as disclosing pulverization of carbonaceous material before graphitizing. However, this would destroy the teachings of the applied Japanese patent application for its intended purpose, as a carbon fiber. Since the teachings of Ryu, et al., as applied by the Examiner, would destroy the applied Japanese patent document for its intended purpose, clearly this combination of teachings as applied by the Examiner is improper. See In re Ratti, 123 USPQ 349 (CCPA 1959).

In any event, even assuming, arguendo, that the teachings of Ryu, et al. were properly combinable with the teachings of the other references as applied by the Examiner, such combined teachings would have neither disclosed nor would have suggested the presently claimed invention, including specific components of the pitch composition, and amounts thereof, and advantages achieved thereby, as discussed previously.

The contention by the Examiner in the third paragraph on page 7 of the Office Action mailed October 14, 2004, is noted. It must be emphasized that Matsumoto discloses forming a fiber. It is respectfully submitted that such disclosure of forming a fiber would have taught away from pulverizing, such pulverizing destroying Matsumoto for its intended purpose. In view thereof, the combining of teachings of references as applied by the Examiner, including the pulverizing, is clearly improper. See In re Ratti, supra.

The contention by the Examiner in the first full paragraph on page 8 of the Office Action mailed October 14, 2004, that Orac, et al. discloses a method of


producing a pitch containing substantially no QI, is noted. It is clear therefrom that the Examiner is using bits and pieces of the teachings of the applied references, without considering the teachings of these references as a whole, it again being emphasized that Orac, et al. discloses a process for converting coal tar containing quinoline-insoluble particles to produce a substantially QI-free coal tar pitch for production of graphite electrodes for the steel industry. Taking the teachings of Orac, et al. as a whole, as required under 35 USC §103, it is respectfully submitted that one of ordinary skill in the art concerned with in Matsumoto, directed to formation of carbon fibers, would not have looked to the teachings of Orac, et al.

In view of the foregoing comments and amendments, entry of the present amendments, and reconsideration and allowance of all claims presently in the application, are respectfully requested.

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Respectfully submitted,

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